



## Superconductivity Centennial Conference

Irreversibility lines and anomalous Meissner effect in  
 $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$  superconducting crystalsO. F. de Lima<sup>a,\*</sup>, R. L. de Almeida<sup>a</sup>, T. M. Garitezi<sup>a</sup>, C. Adriano<sup>a</sup>, P. F. S. Rosa<sup>a</sup>,  
P. G. Pagliuso<sup>a,b</sup><sup>a</sup>*Instituto de Física Gleb Wataghin, Unicamp, 13083-859, Campinas, SP, Brasil*<sup>b</sup>*Department of Physics and Astronomy, University of California Irvine, Irvine, CA - 92697-4575, USA*

---

**Abstract**

Magnetization curves were measured in  $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$  crystals grown by an indium flux method. A very different vortex dynamics behavior was found when comparing results for crystals in the underdoped and slightly above the optimal doped regions. The irreversibility line is closer to the upper critical field line in the latter case, which indicates a much strong pinning of vortices for the near optimally doped crystal. Also, field cooled magnetization curves revealed an anomalous Meissner effect, where a diamagnetic response increases with the applied field, in both crystals. Possible mechanisms involving vortex pinning at interfaces or by local magnetic ions are presented.

© 2012 Published by Elsevier B.V. Selection and/or peer-review under responsibility of the Guest Editors.

Open access under [CC BY-NC-ND license](#).

**Keywords:** FeAs-based superconductors;  $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$  crystals; Indium-flux method; Irreversibility line; Anomalous Meissner effect

---

**1. Introduction**

The study of layered iron-based superconductors over the last three years has brought huge interest, mainly due to an unusual interplay between superconductivity (SC) and magnetism in these materials [1,2]. Besides this basic aspect, these materials also present relatively high values for the superconducting critical temperature ( $T_c$ ), critical current density and upper critical field ( $H_{c2}$ ), which make them attractive for applications [3-5]. Studies on cobalt-doped  $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$  have received especial attention [6,7] mainly because this system is helpful to study the relationship between structural/magnetic phase

---

\* Corresponding author.

E-mail address: [delima@ifi.unicamp.br](mailto:delima@ifi.unicamp.br)

transitions and SC. Also because Co-doping is very convenient for crystal growth, in view of favorable materials processing parameters, leading to relatively homogeneous samples.

The temperature-composition ( $T$ - $x$ ) phase diagram of  $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$  show successive phase transition lines [8,9]; the first is a structural transformation (tetragonal to orthorhombic) at  $T_S$  and the second is an antiferromagnetic ordering (AFM) of the Fe ions at  $T_M$ , such that  $T_S > T_M$ . A third dome-like transition line is where SC appears, at low  $T$ , underneath the regions of AFM/orthorhombic ( $x < x_{\text{op}}$ ) and tetragonal ( $x > x_{\text{op}}$ ) phases, where  $x_{\text{op}}$  is the optimal doping giving the highest  $T_c$ . While there is a consensus that SC resides in a tetragonal structure for  $x > x_{\text{op}}$ , for  $x < x_{\text{op}}$  it is still debatable whether there is coexistence or competition between AFM and SC [2].

We have studied structural, thermal, electrical and magnetic properties of high quality single crystals of composition  $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$  (FeAs-122), grown with an In-rich flux method as described elsewhere [10]. In this paper, we present only results of magnetization measurements in underdoped and near optimally doped crystals, which allow us to discuss the irreversibility lines behavior and an intriguing anomalous Meissner effect.

## 2. Experimental methods

A great number of plate-like crystals were obtained in several runs, for different Co contents, with the larger ones having typical dimensions of 2.5 mm×1.5 mm×0.05 mm. Small amounts of crystals, for selected Co contents, were milled into fine powders and used to obtain X-ray diffraction patterns at room temperature (not shown here). The pure FeAs-122 phase was confirmed and the only impurity detected was indium, deposited in the form of small spots on the surface of some crystals.

Magnetization data were taken with  $H$  parallel to the crystal  $ab$  plane. The measurements were done in a SQUID magnetometer, MPMS-7T, and in a Physical Properties Measurement System, PPMS-14T, both machines from Quantum Design.

## 3. Results and discussion

A large number of  $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$  single crystals were obtained and characterized. Here, we present results for two samples; one in the underdoped region with  $x = 0.03$  and the other around the optimally doped region, with  $x = 0.08$ . The superconducting transition temperature at the onset point and transition width were found to be 20.1 K and 1.8 K for the crystal with  $x = 0.03$ , while for  $x = 0.08$  these values were 20.9 K and 1.1 K.

Fig. 1(a) ( $x = 0.03$ ) and Fig. 1(b) ( $x = 0.08$ ) present magnetization data that reveal paramagnetic and diamagnetic backgrounds, respectively. Curiously the FCC curves in both cases show an enhanced diamagnetic response at low  $T$  when  $H$  is increased. This behavior can be seen more clearly in Figs. 2(a) ( $x = 0.03$ ) and Fig. 2(b) ( $x = 0.08$ ), where the same data are plotted without the normal state magnetic background. In case of Fig. 2(a) suitable straight lines were subtracted from the original data and in case of Fig. 2(b) the original curves were shifted vertically in order to collapse them into the normal base line at  $T \approx 28$  K. In this latter case a bump appears around  $T_c$ , most possibly due to the Curie-Weiss behavior of paramagnetic Fe ions. The inset of Fig. 2(b) displays isothermal FCC magnetization ( $M_{\text{FCC}}$ ) as a function of  $H$ , whose values were obtained at the crossing of the  $M_{\text{FCC}}$  curves with vertical lines passing at

$T = 5$  K and 10 K. In the inset, solid squares (5K) and circles (10 K) refer to the near optimally doped crystal, while solid up triangles (5K) refer to the underdoped crystal.

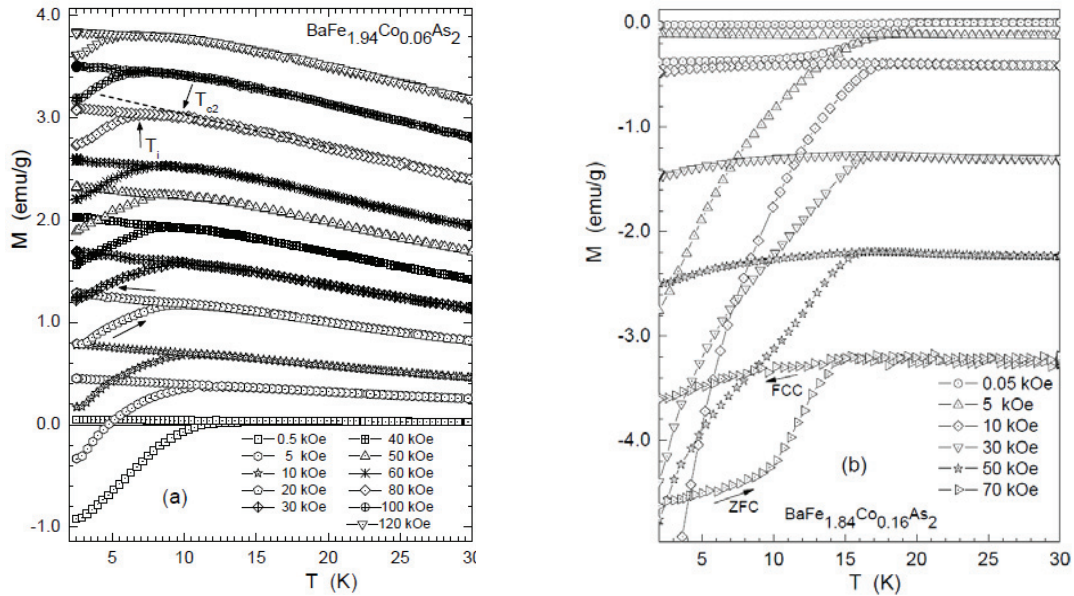


Fig. 1. Magnetization curves as a function of temperature measured for several magnetic fields: (a) for the underdoped crystal; (b) for the near optimally doped crystal. Definitions for the irreversibility point ( $T_i$ ) and onset of transition ( $T_{c2}$ ) are shown in (a).

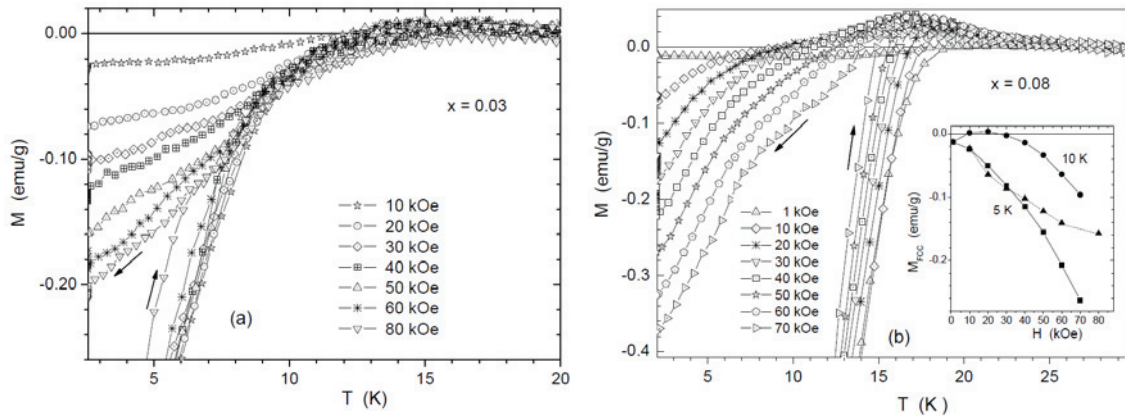


Fig. 2. Magnetization curves as a function of temperature without the normal state magnetic background: (a) for the underdoped crystal; (b) for the near optimally doped crystal. The inset of (b) shows FCC magnetization ( $M_{\text{FCC}}$ ) as a function of  $H$ , whose values were taken at the crossing of the  $M_{\text{FCC}}$  curves with vertical lines passing at  $T = 5$  K and 10 K. Solid squares (5K) and circles (10 K) refer to the near optimally doped crystal, while solid up triangles (5K) refer to the underdoped crystal

The increase of diamagnetic response for higher  $H$ , when FCC measurements are taken, is unusual and different from the regular Meissner effect. Further, this seems to be an intrinsic feature of the FeAs-122 superconductors, since a similar anomalous Meissner effect has already been reported [11] for optimally doped  $\text{BaFe}_{1.85}\text{Co}_{0.15}\text{As}_2$  and  $\text{Ba}_{0.6}\text{K}_{0.4}\text{Fe}_2\text{As}_2$  crystals grown by self flux method.

One possible explanation for the observed anomalous Meissner effect could be related to the occurrence of disordered magnetic moments, produced mainly by Fe ions [11]. In this case, an applied magnetic field could then align these moments, decreasing their efficiency as pair breakers through a possible mechanism that requires spin flipping [12]. Therefore SC could become enhanced, with a stronger diamagnetic response, whenever  $H$  is raised. Interestingly the smaller anomalous effect observed in the underdoped crystal, as shown by the up triangles in the inset of Fig. 2(b), corroborates this idea. Since at  $x = 0.03$  most of the Fe ions are expected to have long range antiferromagnetic order [1,2], they thus could not be participating in the suggested pair breaking mechanism.

Another interesting result arises from the strong difference between the irreversibility lines measured in our two crystals (Fig. 3). Notice that the irreversibility points ( $H_i$ ,  $T_i$ ) were determined at the separation between ZFC and FCC magnetization curves, as exemplified for  $H_i = 80$  kOe in Fig. 1(a). The temperature  $T_{c2}$ , for the onset of the bulk superconducting transition ( $H = H_{c2}$ ), is also defined in that same plot at the point of departure between the magnetization curve and the linear extrapolation of the magnetic normal state base-line.

Fig. 3 shows a distinct vortex dynamics behavior between the two samples. The irreversibility line for the underdoped crystal ( $x = 0.03$ ) is strongly shifted to lower temperatures when compared to the  $H_{c2}$  line position, while this difference is much smaller for the near optimally doped crystal ( $x = 0.08$ ). For instance, these shifts at  $H \approx 5$  T are about 1 K and 8 K, respectively for  $x = 0.08$  and  $x = 0.03$ . This indicates a stronger pinning of vortices in the first case, for the crystal around the optimal doping region.

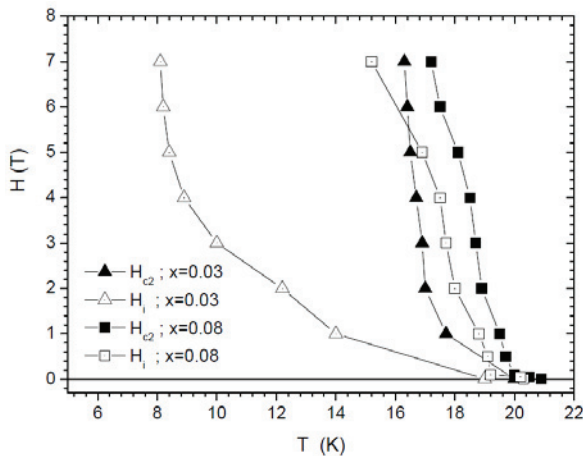


Fig. 3. Irreversibility ( $H_i$ ; open symbols) and upper critical field ( $H_{c2}$ ; solid symbols) lines for the underdoped (up triangles) and near optimally doped (squares) crystals.

Critical current density evaluations for crystals grown in FeAs flux [4] have indicated a higher intrinsic vortex pinning in the optimally doped region, consistent with our findings. A likely reason for that could be associated to an increase of domain boundaries (twins) concentration in the orthorhombic phase, when the Co content approaches the optimal doping. The occurrence of these extended structural interfaces could then produce strong vortex pinning regions. It is worth noting that these twins apparently do not produce granularity effects, which are very common in the high- $T_c$  copper-oxides [13-16].

The occurrence of higher pinning strength, near and above the point of optimal doping, could also be connected to a possible magnetic pinning mechanism caused by the attractive interaction between local magnetic moments and vortex lines [13]. Actually, this would be consistent with a magnetic pair breaking mechanism suggested before, in an attempt to explain the observed anomalous Meissner effect. However, much work is still needed in order to test and advance all these interesting possibilities.

#### 4. Conclusions

Single crystals of  $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ , with  $0.03 < x < 0.10$ , were grown using a new In-flux method. Here, we presented results for two samples; one in the underdoped region ( $x = 0.03$ ), and the other near the optimally doped region ( $x = 0.08$ ).

An anomalous Meissner effect, where the diamagnetic response increases with the applied magnetic field, was observed through field cooled magnetization curves measured in both crystals. This is in disagreement with the usual Meissner effect and seems to be an intrinsic feature of FeAs-122 superconductors. One possible explanation is that an applied magnetic field could align disordered local moments (mainly from Fe ions), decreasing their efficiency as Cooper-pair breakers, through a mechanism that requires spin flipping.

The irreversibility line for the underdoped crystal was found to be strongly shifted to lower temperatures with respect to the  $H_{c2}$  line, while this shift is much smaller for the near optimally doped crystal. This indicates a much strong pinning of vortices for crystals around the optimal doping region. The occurrence of higher density of twin planes in this region could perhaps be the reason for strong vortex pinning at these extended interfaces. It could also be that magnetic pinning, due to attractive interaction between magnetic moments and vortex lines, is operating. This latter idea is very interesting and consistent with the magnetic pair breaking mechanism, suggested to explain the observed anomalous Meissner effect.

#### Acknowledgements

We acknowledge the support from Brazilian science agencies FAPESP (Fundação de Amparo a Pesquisa do Estado de São Paulo) and CNPq (Conselho Nacional de Desenvolvimento Científico e Tecnológico).

#### References

- [1] J. Paglione and R. L. Greene, *Nature Physics* **6** (2010) 645.
- [2] F. Wang and D.-H. Lee, *Science* **332** (2011) 200.

- [3] A. Yamamoto, J. Jaroszynski, C. Tarantini, L. Balicas, J. Jiang, A. Gurevich, D. C. Larbalestier, R. Jin, A. S. Sefat, M. A. McGuire, B. C. Sales, D. K. Christen and D. Mandrus, *Appl. Phys. Lett.* **94** (2009) 062511.
- [4] R. Prozorov, M. A. Tanatar, N. Ni, A. Kreyssig, S. Nandi, S. L. Bud'ko, A. I. Goldman and P. C. Canfield, *Phys. Rev. B* **80** (2009) 174517.
- [5] D. S. Inosov, T. Shapoval, V. Neu, U. Wolff, J. S. White, S. Haindl, J. T. Park, D. L. Sun, C. T. Lin, E. M. Forgan, M. S. Viazovska, J. H. Kim, M. Laver, K. Nenkov, O. Khvostikova, S. Kühnemann and V. Hinkov, *Phys. Rev. B* **81** (2010) 014513.
- [6] A. S. Sefat, R. Jin, M. A. McGuire, B. C. Sales, D. J. Singh and D. Mandrus, *Phys. Rev. Lett.* **101** (2008) 117004.
- [7] P. C. Canfield and S. L. Bud'ko, *Annu. Rev. Condens. Matter Phys.* **1** (2010) 27.
- [8] N. Ni, M. E. Tillman, J.-Q. Yan, A. Kracher, S. T. Hannahs, S. L. Bud'ko and P. C. Canfield, *Phys. Rev. B* **78** (2008) 214515.
- [9] J.-H. Chu, J. G. Analytis, C. Kucharczyk and I. R. Fisher, *Phys. Rev. B* **79** (2009) 014506.
- [10] T. M. Garitezi et al. (to be published).
- [11] R. Prozorov, M. A. Tanatar, B. Shen, P. Cheng, H.-H. Wen, S. L. Bud'ko and P. C. Canfield, *Phys. Rev. B* **82** (2010) 180513.
- [12] R. T. Gordon, H. Kim, M. A. Tanatar, R. Prozorov and V. G. Kogan, *Phys. Rev. B* **81** (2010) 180501.
- [13] B. Blatter, M. V. Feigel'man, V. B. Geshkenbein, A. I. Larkin and V. M. Vinokur, *Rev. Mod. Phys.* **66** (1994) 1125.
- [14] F. M. Araujo-Moreira, W. A. Ortiz and O. F. de Lima, *Physica C* **311** (1999) 98.
- [15] V. P. S. Awana, O. F. de Lima, S. K. Malik, W. B. Yelon and A. V. Narlikar, *Physica C* **314** (1999) 93.
- [16] L. Klein, E. R. Yacoby, Y. Yeshurun, A. Erb, G. M.-Vogt, V. Breit and H. Wühl, *Phys. Rev. B* **49** (1994) 4403.